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September 13, 2006

Journal of Alloys and Compounds

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## Density changes in Ga-stabilized delta-Pu, and what they mean

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### Abstract

Ga-stabilized  $\delta$ -Pu undergoes small changes in density with time. These have been associated with four different causes: an initial reversible expansion that saturates after a short time; a continuous change that can be attributed to the in-growth of helium and actinide daughter products from the radioactive decay of plutonium; possible void swelling; and phase instability. We review our present understanding of these processes and evaluate their contributions to density changes. It is shown that the initial transient expansion is intimately connected with the metastability of the  $\delta$ -phase at ambient temperature.

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## 1. Introduction

Density measurements of Plutonium-Gallium alloys have always been conducted as a means to assure that the material has the intended chemical composition, the appropriate phases, and the desired metallurgical treatment. As a potential indicator of aging due to radioactive decay, however, density and dimensional measurements have only recently been performed and analyzed in any depth. Also recently, the length of specimens with higher concentrations of the short-living isotope  $^{238}\text{Pu}$  has been, and continues to be continuously monitored. As a result of these recent data and of theoretical and computational studies, the earlier picture [1], illustrated schematically in Fig. 1, has undergone significant changes. Nevertheless, the original division into four major mechanisms of density changes has remained a useful guide.

According to that division, a short transient expansion occurs initially in both lattice parameter and specimen length, and it saturates quickly. Its main feature is that it can be reversed by heating the material to 150 to 200 C. It was originally believed to be due to defect clusters, remnants of the large collision cascades that the U recoils produce. However, the most recent atomistic simulations of collision cascades in Ga-stabilized  $\delta$ -Pu, to be discussed in Section 2, reveal an atomic structure of collision cascades radically different from those in other fcc metals. For this reason and others, we present in Section 3 another explanation for the transient, reversible expansion.

Helium generated in the  $\alpha$ -decay of plutonium accumulates in the form of bubbles [2]. This gives rise to a continuous, volumetric expansion, but to no significant change in lattice parameter. Since the helium content increases almost linearly with time or age, it was originally assumed that helium bubble swelling also increases linearly with time, and

that the initial swelling rate can be extrapolated. More recent theoretical studies summarized in Section 4 showed, however, that actinide daughter products, namely Am, U, and Np, lead to changes in lattice parameter and length. Their combined contributions results in an irreversible expansion that is non-linear for weapons-grade material, but nearly linear for  $^{238}\text{Pu}$ -enriched alloys.

Void swelling is another phenomena originally expected to occur in Ga-stabilized  $\delta$ -Pu self-irradiated at ambient temperatures [1]. So far, however, voids have not been observed in aged specimens examined by transmission electron microscopy, although high densities of nano-meter sized helium bubbles are regularly found in such specimens. Our theoretical and computational studies on void nucleation and swelling are summarized in Section 5, and they indicate that the propensity for void swelling is very low for Ga-stabilized  $\delta$ -Pu.

The final source of density changes is associated with phase changes, principally with the possible instability of the Ga-stabilized  $\delta$ -Pu phase. It is now known that this phase is in fact thermodynamically metastable at ambient temperatures [3]. The new explanation of the transient, reversible expansion turns out to be closely related to the question of phase stability, and as argued in Section 6, it provides surprising, though circumstantial evidence that self-radiation damage is a key factor in maintaining the Ga-stabilized  $\delta$ -Pu phase in its metastable state.

## **2. The Primary Displacement Damage**

When a Pu atom decays, four radiation damage processes are enacted. First, the  $\alpha$ -particle with an initial kinetic energy of 5 MeV traverses a distance of about 10  $\mu\text{m}$ ,

causes ionization and electron excitations along its way, captures electrons as it slows down, creates some Frenkel pairs, and at the end of its path creates a small collision cascade with about 200 Frenkel pairs. Second, when it comes to rest, it is then a helium atom, most likely occupying one of the vacancies it created: it has become a substitutional impurity in  $\delta$ -Pu. Its subsequent aggregation with other helium atoms leads to helium bubble formation [4].

Third, the Pu atom that decayed turns into an U atom with an initial kinetic energy of about 85 keV. It dissipates 95 % of its energy by dislodging thousands of other Pu atoms from their normal positions in the crystal lattice, thereby creating a large collision cascade.

Forth, the generation of U and other actinides changes the alloy composition and hence, the properties of the material.

Collision cascades are studied by classical molecular dynamics methods using empirical interatomic potentials. Only recently has it become feasible to study the large U collision cascades with the most advanced supercomputers, such as BlueGene/L. Fig. 2 shows three snapshots of such a collision cascade at three different times after the  $\alpha$ -decay.

This particular simulation involved a 85 keV recoil producing a cascade in a supercell containing 16 million Pu atoms in an fcc crystal held at 600K. The simulation has been carried out for over 1 ns, using the Modified Embedded Atom Method (MEAM) interatomic potentials of Baskes [5]. In Figure 2, the red atoms are “vacancies” and the white atoms are “self-interstitials”. These defects are determined by examining the occupancy of the Wigner-Seitz cells of the initial undisturbed lattice. Vacancies and self-interstitials are defined as locations of Wigner-Seitz cells containing zero or

two atoms, respectively, at any moment in time. However, the atomistic structure of the collision cascade in  $\delta$ -Pu resembles that of an amorphous region, and an identification of “vacancies” and “self-interstitials” is realistically not possible, hence the quotation marks. These irregular Pu atoms rearrange and slowly restore to a more regular structure close to a crystal lattice structure. What is unique for  $\delta$ -Pu, is that this restoration of the fcc crystal structure takes place very slowly. In other fcc metals, it takes only about 100 to 200 ps for a final state to be reached. In contrast, as shown in Fig. 3, the number of “Frenkel pairs” diminishes very slowly even at the elevated temperature of 650 K, chosen to accelerate the recovery. These initial studies indicate that the disordered atomic structure of a collision cascade in  $\delta$ -Pu persists for nearly a  $\mu$ s at 650 K, and perhaps several ms at ambient temperatures. Furthermore, what remnant damage remains, if any, is still uncertain and must await longer computer runs.

Further studies underway indicate that Ga in the Pu lattice has the effect of increasing the recovery time of the amorphous region back to the fcc lattice.

### **3. The Transient, Reversible Expansion**

The volumetric expansion of Ga-stabilized  $\delta$ -Pu can be observed both via length change measurements as well as by x-ray diffraction analysis. Fig. 4 shows a composite plot containing of one dilatometry curve of a sample enriched with 0.75 %  $^{238}\text{Pu}$  [6] and held at 35 C together with lattice parameter measurements on materials with nominal (about 0.02%) and enhanced  $^{238}\text{Pu}$  content [7, 8]. The time axis is normalized such that a dose of 1 dpa corresponds to 10 years of age for weapons-grade material. The transient expansion during the first two years is followed by a gradual, slow expansion. What is

most unusual, however, is the fact that the length change is less than the lattice parameter change, i.e.,  $\Delta l / l < \Delta a / a$ . If the dimensional changes were due to defect clusters from the displacement damage, the length change should either be equal to or greater than the lattice parameter change.

To explain this anomaly, Wolfer et al. [9] proposed that the transient expansion arises from the short-range ordering of Ga or, what amounts to the same, from the incipient precipitation of the  $\zeta'$ -phase, the  $L1_2$  compound  $\text{Pu}_3\text{Ga}$ . As seen in Fig. 5 [10], this phase occupies a smaller volume per atom than the Ga-stabilized  $\delta$ -Pu phase. At the same time, the incipient precipitation of the  $\zeta'$ -phase depletes the Ga-stabilized  $\delta$ -Pu phase of gallium atoms, thereby increasing its lattice parameter. X-ray diffraction experiments only reveal the lattice parameter of the Ga-stabilized  $\delta$ -Pu phase, as the volume fraction and the size of  $\zeta'$ -precipitates is too small. Nevertheless, they do contribute to the overall dimension of a sample. If  $a(x_0)$  denotes the lattice parameter of a sample with a Ga content of  $x_0$  atomic fractions, and  $\Delta a(x_0)$  is the lattice parameter increase of the Ga-stabilized  $\delta$ -Pu phase, then the length change of the sample is given by [9]

$$\frac{\Delta l(x_0)}{l(x_0)} = \frac{\Delta a(x_0)}{a(x_0)} \left[ 1 - \frac{4 a(x_0)}{3\beta} \left( \frac{a_\zeta^3}{a^3(x_0)} - 1 \right) \right] \quad (1)$$

Here,  $\beta$  is the slope of the lattice parameter  $a(x_0)$  for the  $\delta$ -phase, and  $a_\zeta$  is the lattice parameter of the  $\zeta$ -phase, both are shown in Fig. 5. Evaluation of this equation for  $x_0 = 0.02$  reveals that the length change is only 42% of the lattice parameter change.



The time evolution of the lattice parameter change has been measured carefully by Oudot [11], and Fig. 6 shows his results for an alloy with 3.7 at.% Ga. They can be fitted to the equation

$$\Delta a(\tau) = \Delta a_{sat} [1 - \exp(-\tau / \tau_{RID}^{\zeta})] \quad (2)$$

with

$$\tau_{RID}^{\zeta} = 0.022 \pm 0.003 \text{ dpa} \quad (3)$$

Note that a saturation value is reached with no subsequent expansion from helium accumulation, as helium bubbles do not change the lattice parameter, but only the external dimensions of the sample.

The saturation values  $\Delta a_{sat}/a$  obtained by Oudot are shown in Fig. 7 together with the earlier results from Chebotarev et al. [12] as a function of the gallium content. The relatively small values for the change in lattice parameter imply that only a small fraction of gallium is depleted in the Ga-stabilized  $\delta$ -Pu phase and is associated with the incipient  $\zeta'$ -precipitates. We return to this point in Section 6 when discussing the stability of the Ga-stabilized  $\delta$ -Pu phase.

#### 4. Permanent Density Changes from Helium and Daughter Products

The isotopes contained in Pu and their abundances are listed in Table 1. Their

decays not only produce helium, but also the daughters Am, U, and Np. Fig. 8 shows the concentrations of these isotopes and of He in weapons-grade Pu as a function of age. This is compared in Fig. 9 with the concentrations in the  $^{238}\text{Pu}$  enriched

material as a function of the equivalent age. In this latter figure, the concentrations of U and He are nearly identical, and they do not display as separate lines.

Wolfer et al. [13] have carried out a detailed analysis of the volumetric changes produced by the accumulation of helium and of the daughter products. Helium contained in bubbles expands the external volume of  $\delta$ -Pu by about two atomic volumes of Pu for every 5 helium atoms generated. At the same time, however, the same number of actinide daughters U and Np are generated as are He atoms. These elements have smaller atomic volumes than Pu in its  $\delta$ -phase, and about 5 atoms of them occupy the space of just 4 Pu atoms. As a result, the net volumetric expansion from the radioactive decay of 5 Pu atoms is about one atomic volume. However, this simple analysis does not account for the contribution of Am. It is a larger atom than Pu, and about 4 Am atoms occupy the space of 5 Pu atoms in its  $\delta$ -phase. Am is rapidly produced by the  $\beta$ -decay of  $^{241}\text{Pu}$ , but it decays then with a half-life of 458 years into Np. In spite of the low concentrations of Am, its contributions to volume and lattice parameter changes manifest themselves early, and they must be included in the analysis of experimental results and their extrapolation to higher ages.

In fact, in the Figs. 10, 11, and 12, the changes in volume and in lattice parameter exhibit a non-linear behavior after the initial transient, i.e. after about 2 years. This non-linearity is entirely due to the Am build-up and its subsequent decline after about 30 years.

Fig. 10 shows the volumetric expansion for weapons-grade material and the experimental data of immersion density on samples extracted from retired pits. The large scatter in these data is due to the uncertainty in the initial density. Nevertheless, the comparison demonstrates that the theoretical predictions are not in conflict with the data.

A similar comparison for lattice parameter changes is shown in Fig. 11. Again, considering the uncertainty in the measured values, as indicated by one typical error bar, there exist agreement between predictions and experimental results. These predictions are extended to 100 years and displayed in Fig. 12. Because helium bubbles do not contribute to an increase in lattice parameter, and because Am ceases to enlarge it after 30 years, the lattice parameter declines after 30 years, while the external dimensions continue to increase. The initial anomaly, namely that the lattice parameter increase is greater than the length increase, reverses to the normal behavior after 30 years.

Since Am makes a very minor contribution to the dimensional changes of  $^{238}\text{Pu}$ -enriched samples, they expand, after the initial transient, at a constant rate as shown in Fig. 12. This rate is in fact equal to the asymptotic expansion rate that weapons-grade material reach beyond 60 years.

## **5. The Prospect for Void Swelling in Plutonium**

Void swelling is a general phenomenon in metals and alloys subject to displacement damage in a temperature range between 0.3 and 0.65 of the melting point [1]. Apart from the temperature requirement, three other conditions must be satisfied for void swelling to occur. First, not all self-interstitials should recombine with vacancies either within their cascade of origin or subsequently during their migration away from it. Second, during this subsequent diffusion, the self-interstitials should be attracted and captured preferentially at dislocations, so a vacancy surplus can develop, or in other words, a vacancy supersaturation can build up. The latter is a necessary condition for voids to

nucleate. Third, these void nuclei need to be stabilized once nucleated to prevent their collapse into prismatic dislocation loops. This is accomplished by the capture of helium atoms or other impurities that segregate on the void surfaces.

Impurities, minor alloying elements, and small precipitates also enhance the recombination of vacancies and interstitials as they migrate through the crystal lattice; either one or both can be temporarily trapped at these imperfections, thereby enhancing the chance to recombine before being captured at dislocations or voids. Defect trapping dramatically prolongs the incubation time for the onset of void swelling [14], as seen in Fig. 13. While pure copper begins to swell almost at the start of the irradiation, impure and alloyed copper may remain swelling resistant up to 50 dpa.

Weapons-grade plutonium contains plenty of impurities that are expected to greatly enhance recombination. In addition, the amorphous structure of the cascade region also implies that vacancies and interstitials may not exist in it, or that their recombination occurs already within the cascade.

Helium is plentiful and readily available to stabilize voids if they were to nucleate. However, helium bubbles have formed long before voids would have nucleated. They then act as effective recombination centers, and prevent a high vacancy supersaturation to become established. These qualitative considerations indicate that the first condition for void formation is not at all favorable in weapons-grade plutonium.

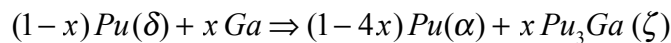
It is the second condition, however, that is not likely to be met in plutonium. A measure for the preferential capture of self-interstitials over the capture of vacancies by dislocations is the so-called bias. It needs to be positive if voids are to nucleate and to

grow. A positive bias results when the absolute value of the interstitial's relaxation volume is larger than the corresponding value for the vacancy.

First-principle calculations have been carried out with the VASP code on self-interstitials and vacancies in a pure  $\delta$ -Pu crystal lattice, allowing for full relaxation of atom positions after introducing the defect. The relaxation volumes so obtained are as follows. The vacancy in  $\delta$ -Pu has a relaxation volume of  $-0.62 \Omega_\delta$ , while the self-interstitial in the  $\langle 100 \rangle$  dumbbell orientation possesses a relaxation volume of  $0.38 \Omega_\delta$  and  $0.63 \Omega_\delta$  in the  $\langle 111 \rangle$  orientation. Here,  $\Omega_\delta$  is the atomic volume of  $\delta$ -Pu. Clearly these results indicate that the bias is not positive, and the second condition for void swelling is not fulfilled. Before a final conclusion regarding void swelling in Pu can be reached, however, calculations for defect relaxation volumes need to be performed for Ga-stabilized  $\delta$ -Pu.

## 6. Stability of Ga-stabilized $\delta$ -Pu

The phase diagram in the region of low gallium content has only recently been finalized with the work of Timofeeva [3], and it is shown in Fig. 14. Accordingly, it is now accepted that Ga-stabilized  $\delta$ -Pu is metastable at ambient conditions. To reach thermodynamic equilibrium would require the following reaction to proceed to completion:



But even a partial transformation with some formation of the  $\zeta'$ -phase will lower the Gibbs free energy of the material. An alternate approach towards equilibrium is the

martensitic transformation to the  $\alpha'$ -phase. However, this phase is also a metastable one, namely an  $\alpha$ -phase supersaturated with gallium. This transformation can occur only at low temperature, or under pressure. At ambient conditions then, the only path to approach equilibrium is by the formation of  $\text{Pu}_3\text{Ga}$ , the  $\zeta'$ -phase; but this requires diffusion of gallium, or at least short-range ordering of gallium. Radiation-enhanced diffusion is probably the mechanism for gallium migration at ambient temperatures.

In Section 3 we have invoked the incipient precipitation of the  $\zeta'$ -phase for the transient expansion that has the peculiar characteristic of creating a length change less than a lattice parameter change. The second characteristic is that it is reversible when heating the material to a temperature above about 150 C. Fig. 15 shows the results of such a recovery experiment by Koslov et al. [15]. A sample which underwent the transient expansion is heated. Up to 100 C, the sample elongates along the red curve by thermal expansion, but then ceases to expand as thermal elongation is compensated by contraction from the thermal dissolution of the  $\zeta'$ -phase. This dissolution increases the gallium content of the  $\delta$ - phase, and restores its lattice parameter as well as the sample length to what it was originally at room temperature. When heated a second time shortly after the first annealing, the sample elongates by thermal expansion only along the blue curve. Note that the two elongation curves are shifted in Fig. 15 to have the same starting point even though the sample had two slightly different lengths.

The interpretation of this recovery experiment is a logical consequence of the incipient precipitation of the  $\zeta'$ -phase and its subsequent dissolution when the temperature is raised into the region of the phase diagram where the  $\delta$ -phase is thermodynamically stable.

What remains to be explained is the cessation of the  $\zeta'$ -precipitation. An increase  $\Delta a$  in the lattice parameter of the Ga-stabilized  $\delta$ -Pu phase implies a certain atomic fraction  $\Delta x$  of gallium being incorporated in the  $\zeta'$ -phase, namely

$$\Delta a = -\beta \Delta x = 0.8794 \Delta x \quad (4)$$

Saturation for  $\Delta a$  implies a saturation for  $\Delta x$ , the gallium depletion of the  $\delta$ -phase. From the results for  $\Delta a_{sat}$  shown in Fig. 7, we obtain with Eq. (4) the final depletion fraction  $\Delta x_{sat}/x$  shown in Fig. 16. Here,  $x$  is the initial gallium content in the  $\delta$ -phase.

A better physical meaning can be gleaned from the differential equation that gives this solution. This equation is

$$\frac{d[\Delta x(\tau)]}{d\tau} = P - \frac{\Delta x(\tau)}{\tau_{RID}^{\zeta}} \quad (5)$$

We can interpret the first term as the precipitation rate of the  $\text{Pu}_3\text{Ga}$  compound and the second as the rate of its destruction by radiation-induced disordering (RID). Then, according to the result stated in equation (3), the half-life of a  $\zeta'$ -precipitate is  $0.022 \ln 2 = 0.015$  dpa.

It becomes now obvious that the new explanation for the reversible transient expansion also provides the key to unlock the mystery of gallium stabilization for the  $\delta$ -phase:

Gallium, being nearly insoluble in all the phases of Pu except in the  $\delta$ - and  $\epsilon$ -phase, restricts the ranges of existence for the  $\beta$ - and  $\gamma$ -phase, and thereby expands it for the  $\delta$ -phase. The extension for the  $\delta$ -phase to lower temperature is just far enough to turn off self-diffusion before leaving the range of thermodynamic stability, as can be seen in Fig.

14. As a result, the Ga-stabilized  $\delta$ -Pu is unable to transform at ambient temperature by thermal diffusion. Radiation-induced diffusion facilitates some migration of gallium and some incipient precipitation of the equilibrium  $\zeta'$ -phase. Fortunately, this ordered  $L1_2$  structure can also be disordered by the large collision cascades that the U recoils produce; atomically disordered gallium on a face-centered cubic lattice of Pu is of course nothing else but the Ga-stabilized  $\delta$ -phase. So in the final analysis, the stabilization of the  $\delta$ -phase is a dynamic process that depends not only on the presence of gallium but also on its continuous redistribution brought about by the self-irradiation of plutonium.

## Acknowledgements

This work was performed under the auspices of the US Department of Energy by the University of California Lawrence Livermore National Laboratory under contract W-7405-Eng-48.

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## Figure Captions

Fig.1. Conceptual behavior of the volumetric changes in plutonium at ambient temperature.

Fig.2. Three snapshots of a 85 keV collision cascade in  $\delta$ -Pu at times 10.45 ps, 55.77 ps, and 2133 ps after the  $\alpha$ -decay event. Red are empty and white are doubly occupied Wigner-Seitz cells. Although they are referred to as “vacancies” and “self-interstitials” and as pair as “Frenkel pairs”, the atomic structure of this cascade is in fact amorphous.

Fig.3. The recovery of the 85 keV collision cascade with time exhibits three stages. The first stage at 15 ps marks the end of the collision sequence. The second and third stages can be fit to  $[18148-10253 \exp(-77.83/t)]$  and  $[14142-1223 t^{0.212}]$ , respectively, where the time is in ps.

Fig.4. Volumetric expansion derived from lattice parameter changes,  $da/a$ , and length changes,  $dl/l$ .

Fig.5. The lattice parameters of Ga-stabilized  $\delta$ -Pu alloys and of  $\text{Pu}_3\text{Ga}$  at ambient temperature.

Fig.6. The transient lattice parameter change for Ga-stabilized  $\delta$ -Pu with 3.7 at.% Ga.

Fig.7. Saturation values for the transient lattice parameter change for different Ga contents.

Fig.8. Concentrations of helium and actinide decay products in weapons-grade plutonium.

Fig. 9. Concentrations of helium and actinide products in weapons-grade plutonium enriched with 7.38 at.% of  $^{238}\text{Pu}$ .

Fig. 10. Comparison of volumetric changes measured by immersion density and theoretical predictions. The maximum and minimum curves correspond to a helium density within bubbles of 2 and 3 helium atoms per missing Pu atom, respectively.

Fig. 11. Comparison of volumetric changes inferred from lattice parameter changes. Diamonds are for weapons-grade material and squares for  $^{238}\text{Pu}$  enriched material. Time is measured in equivalent years, normalized to real time for weapons-grade material.

Fig. 12. Predicted dimensional changes for weapons-grade (WG) material and for  $^{238}\text{Pu}$  enriched material (AA).

Fig. 13. Void swelling in neutron-irradiated Cu and Cu alloys.

Fig. 14. Portion of the Pu-Ga phase diagram .

Fig. 15. Linear thermal expansion of Ga-stabilized  $\delta$ -Pu with and without the recovery of the transient lattice parameter change.

Fig. 16. Fraction of Ga depleted from the  $\delta$ -phase and contained in the  $\zeta'$ -phase.

**Table 1. Radioactive Decay of Plutonium Isotopes**

Parent Isotope	Abundance, %		Decay	Half-life years	Daughter Isotope
	WG	Spiked*			
<sup>238</sup> Pu	0.02	7.38	α	87	<sup>234</sup> U
<sup>239</sup> Pu	93.6	86.2	α	24390	<sup>235</sup> U
<sup>240</sup> Pu	5.9	5.52	α	6580	<sup>236</sup> U
<sup>241</sup> Pu	0.44	0.15	β	14.63	<sup>241</sup> Am
<sup>241</sup> Am			α	458	<sup>237</sup> Np
<sup>242</sup> Pu	0.04	0.057	α	3.8 10 <sup>5</sup>	<sup>238</sup> U

\* WG stands for weapons-grade, and Spiked for material enriched in <sup>238</sup>Pu.

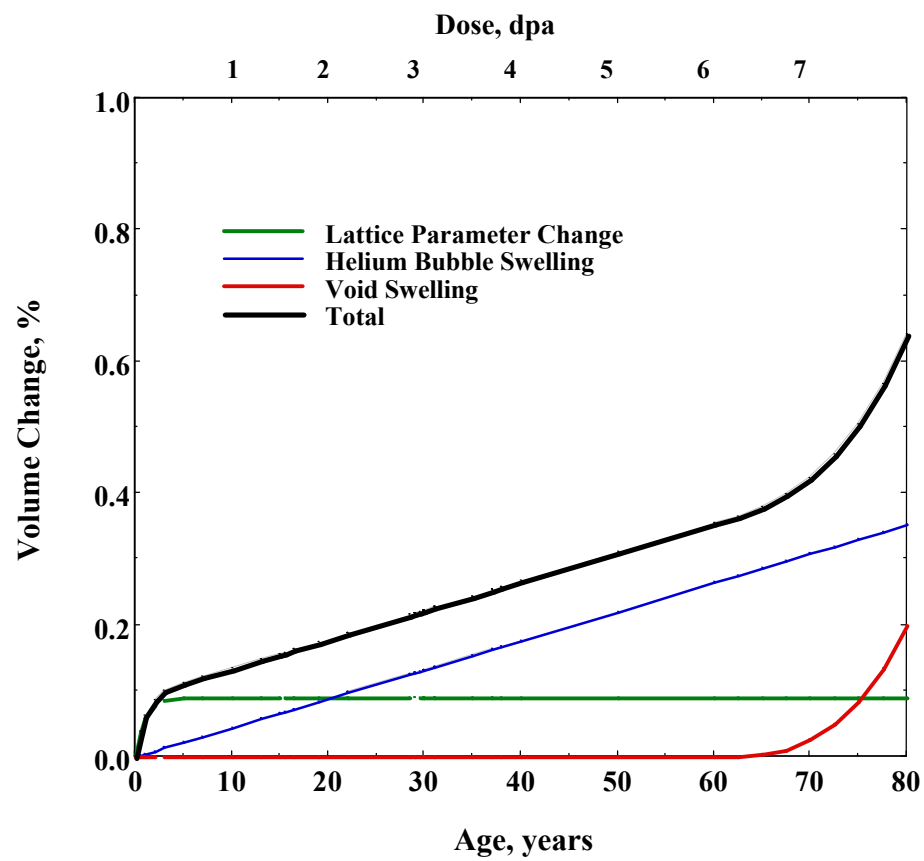
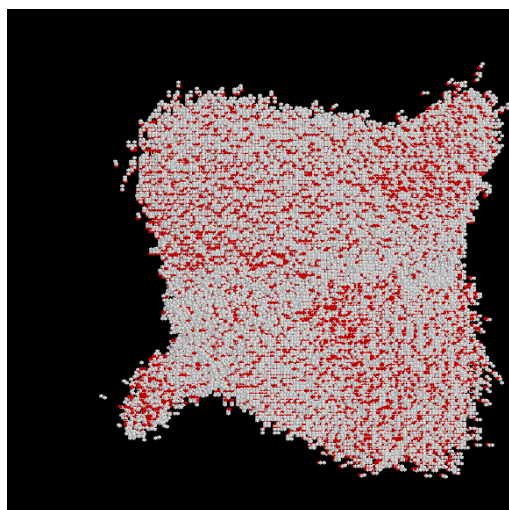
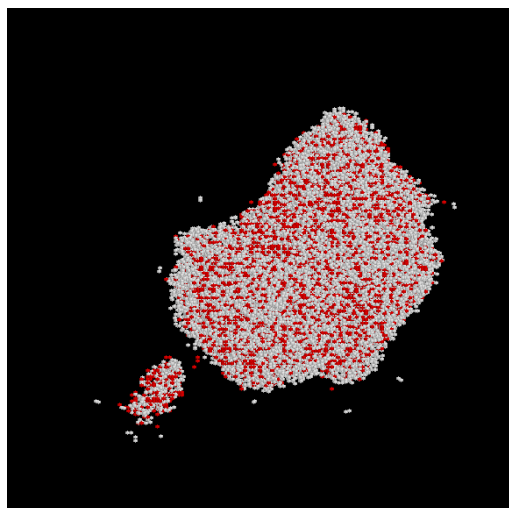


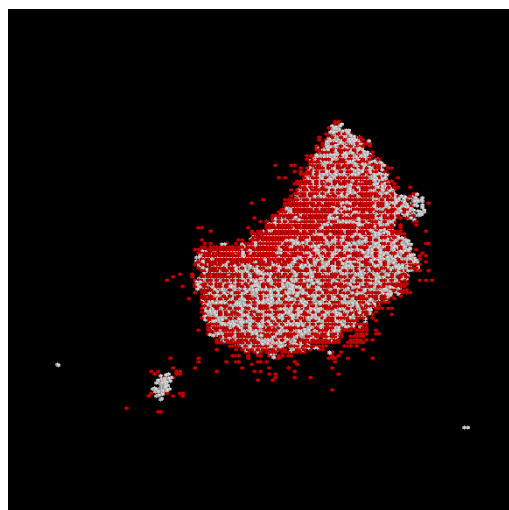
Fig. 1



10.45 ps



55.77 ps



2133 ps

Fig. 2

“ Frenkel Pairs “

QuickTime™ and a  
QuickTime decompressor  
are needed to see this picture.

Time, ps

Fig. 3

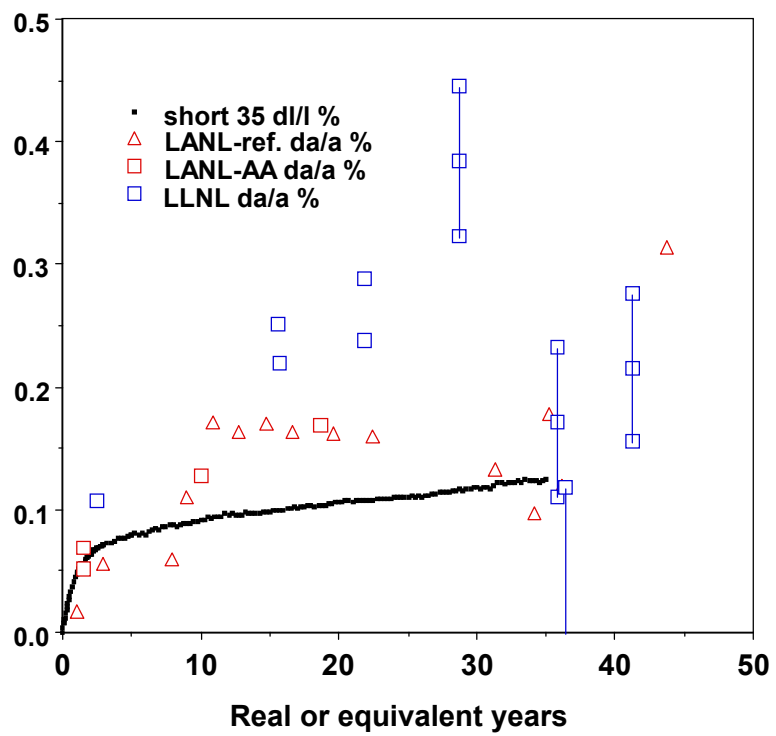


Fig. 4



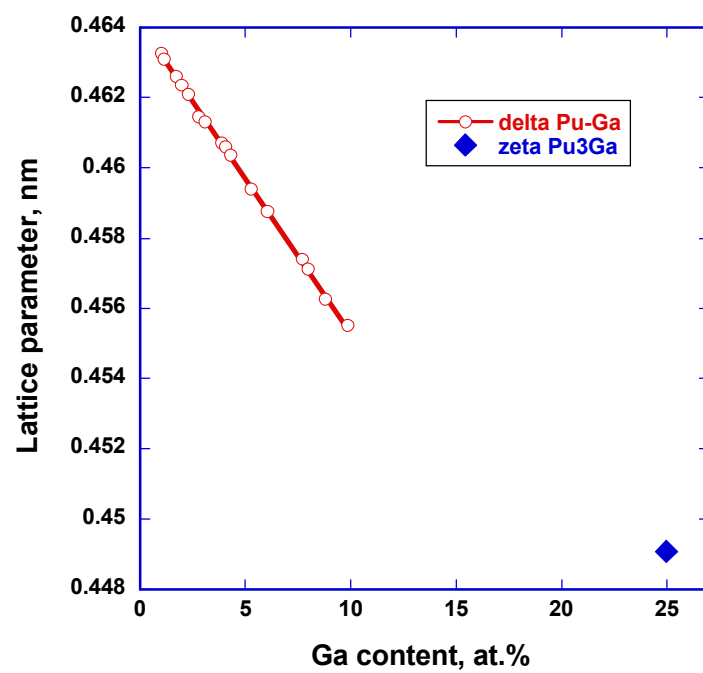


Fig. 5

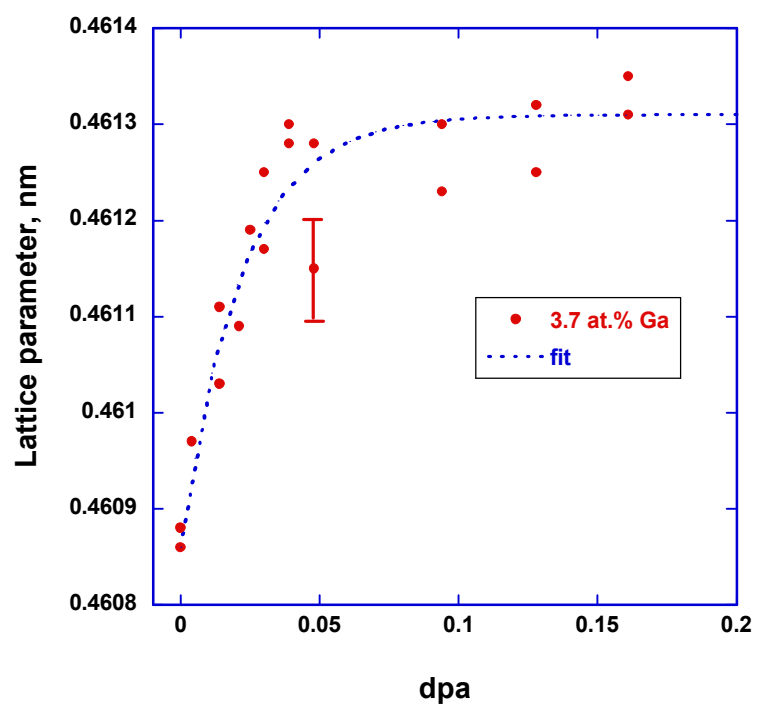


Fig. 6

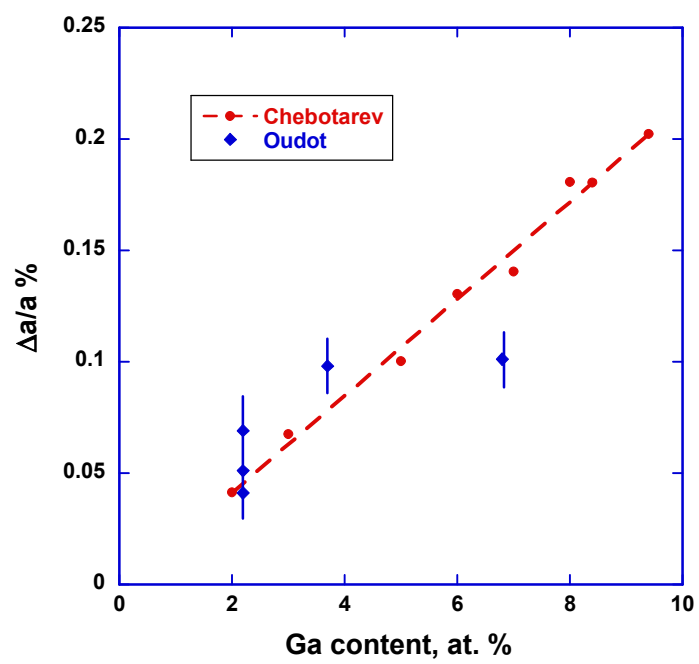


Fig. 7

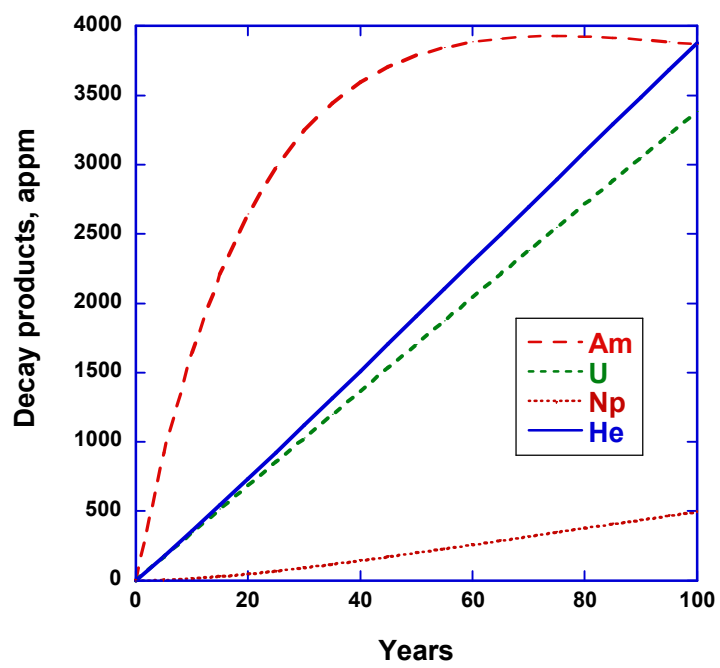


Fig. 8

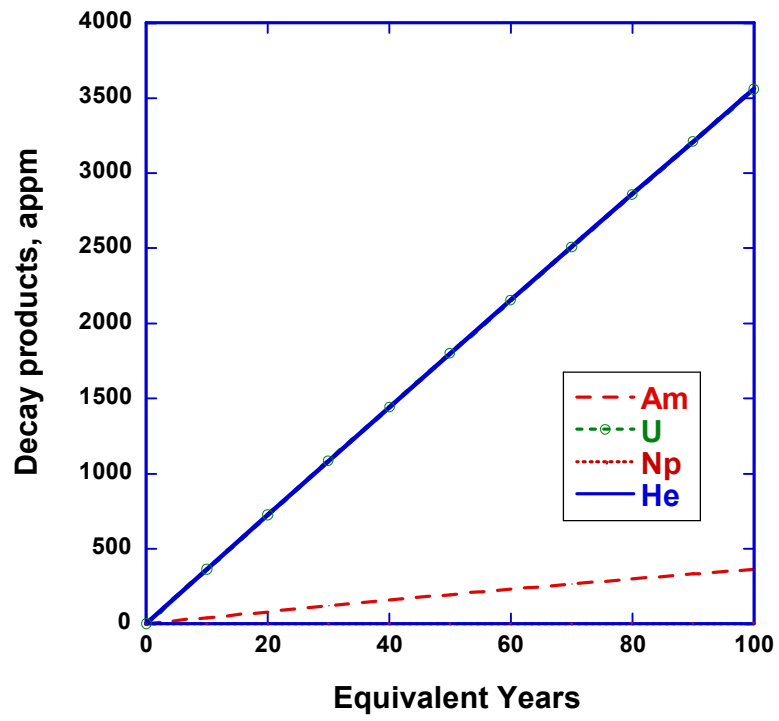


Fig. 9

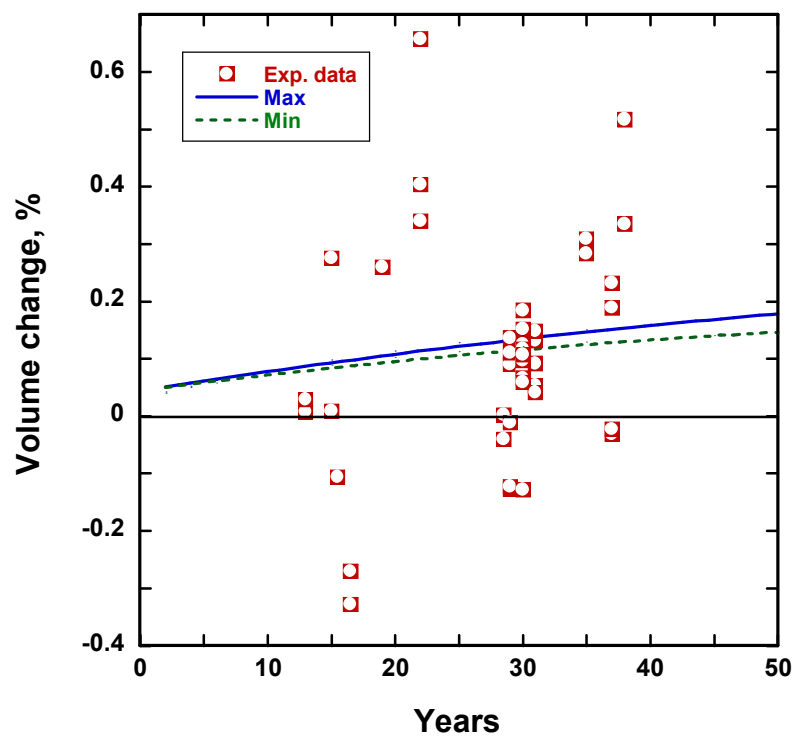


Fig. 10

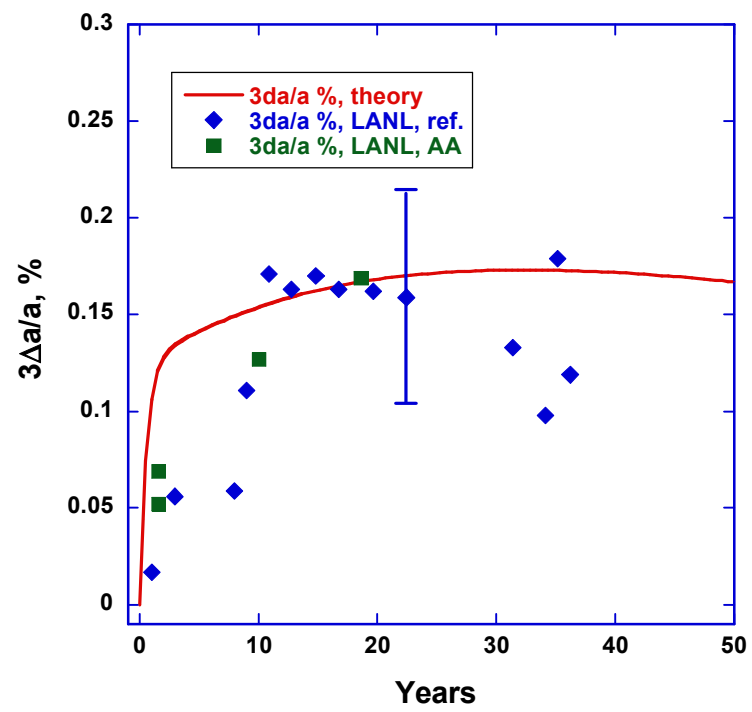


Fig. 11

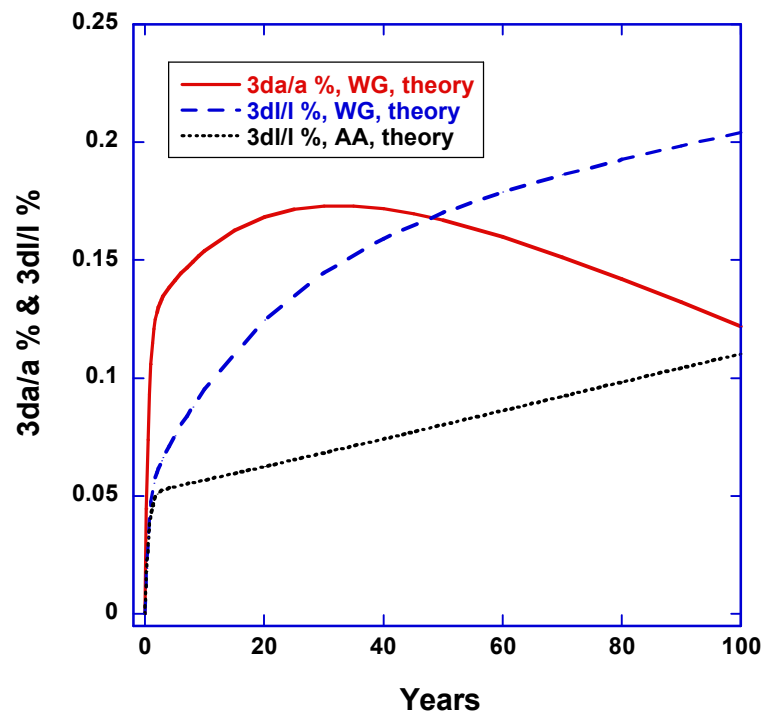


Fig. 12



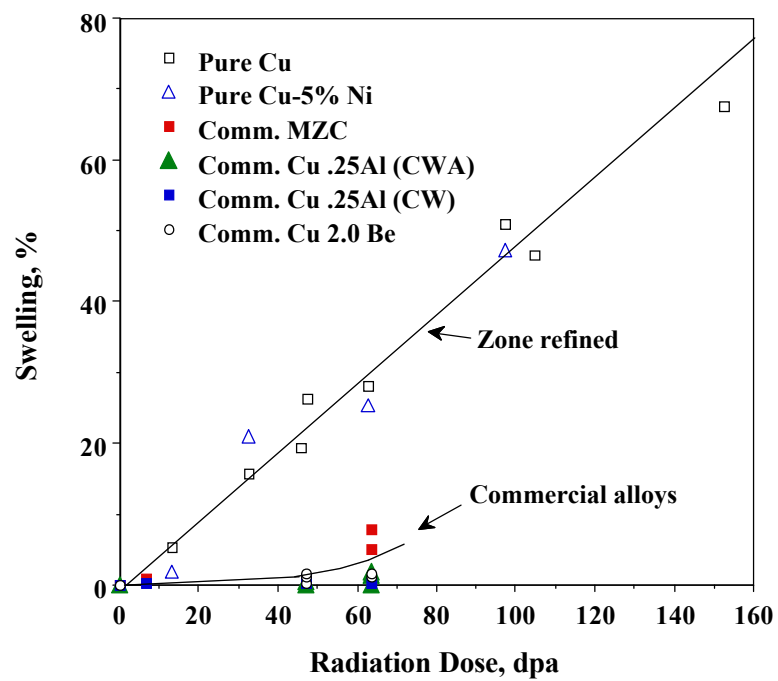


Fig. 13

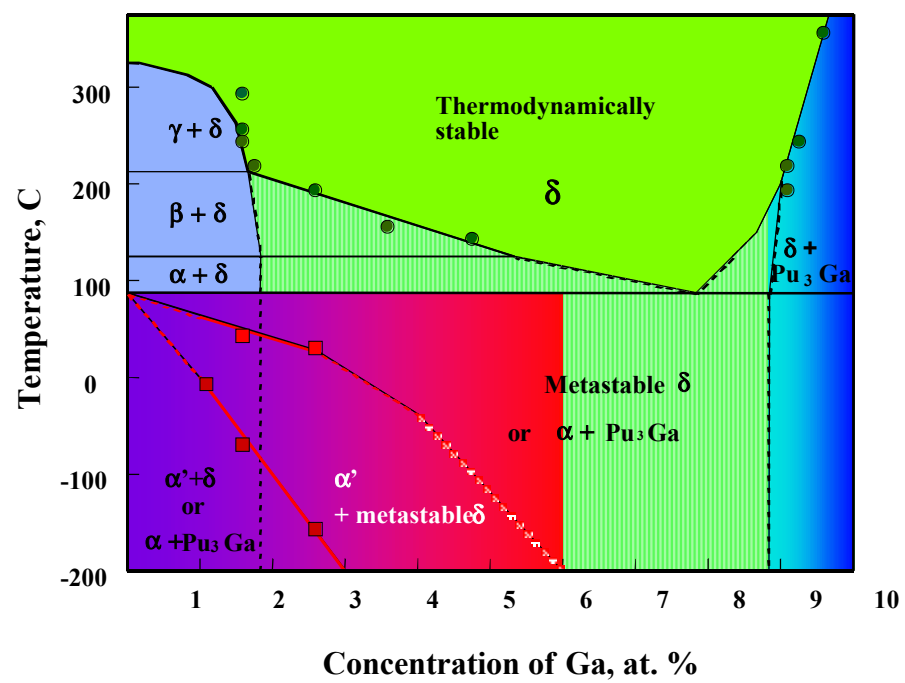


Fig. 14

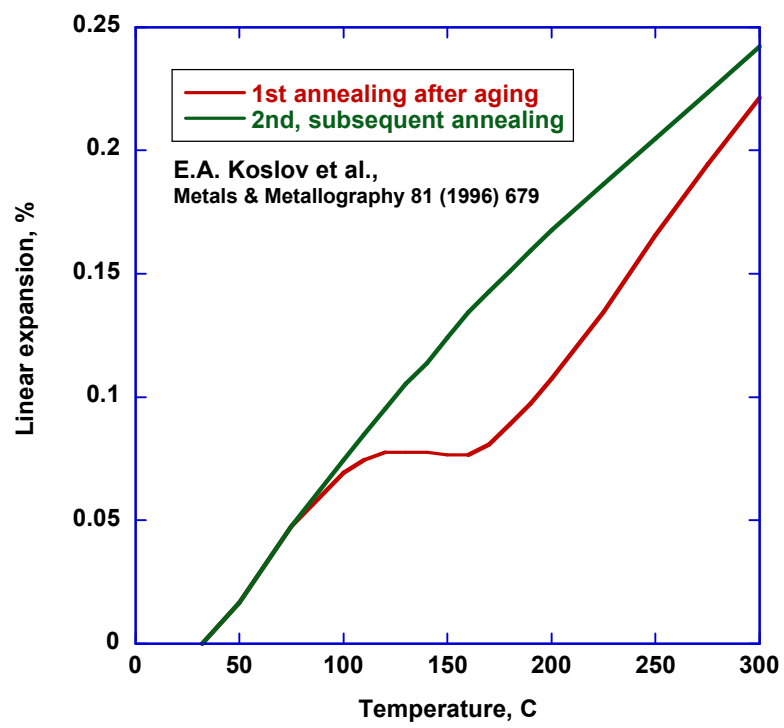


Fig. 15

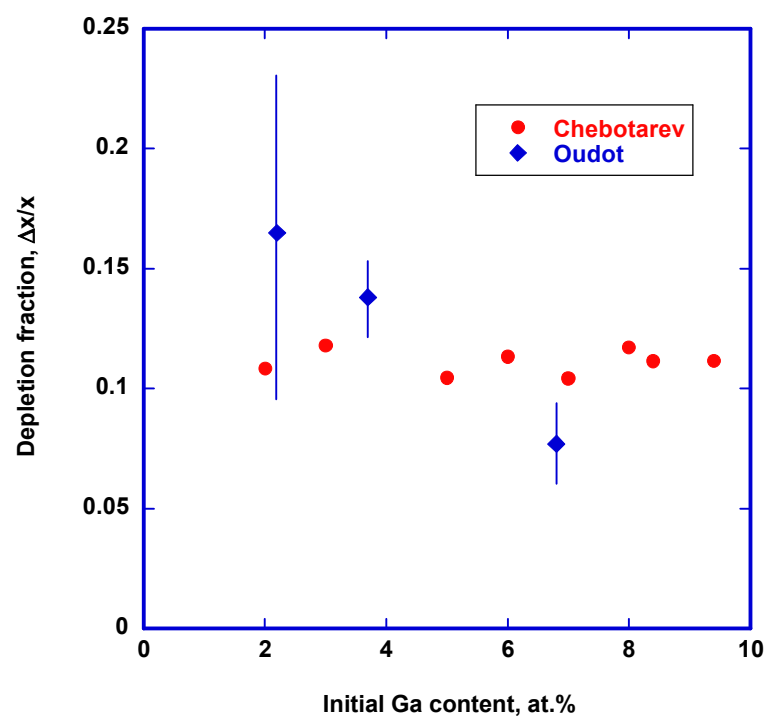


Fig. 16

